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THERMODYNAMIC PROPERTIES OF ETHANOL AT ATMOSPHERIC PRESSURE

T. S. Khasanshin and A. A. Aleksandrov

UDC 536.7:547.262

Published data on density, speed of sound, and isobaric specific heat in liquid ethanol are generalized, and the values of isochoric specific heat, adiabatic and isothermal compression coefficients, and enthalpy are calculated for the temperature range 159.05-351.44°K.

The thermodynamic properties of pure (absolute) ethanol at atmospheric pressure have been studied quite thoroughly. The accumulated experimental material was systematized and partially generalized in [1-7]. The generalization was accomplished in the form of recommended interpolation equations or data tables. As a rule, recommended values were provided for temperatures above 273°K. However, modern methods of constructing thermodynamic property tables for liquids, in particular, those based on data from acoustical measurements, require the representation of a number of thermodynamic properties (ρ, W, and C_p) at atmospheric pressure as analytic expressions. Presently available handbooks and monographs either lack such equations or offer ones limited to a narrow temperature range, which are furthermore insufficiently precise in a number of cases.

It is obvious that the accumulated experimental data on the properties of ethanol and the generalizations thereof are not exhaustive, since new experimental data has been published, which with consideration of previous material makes possible a refinement and generalization

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Mogilev Technological Institute. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 47, No. 3, pp. 411-418, September, 1984. Original article submitted April 25, 1983.

0022-0841/84/4703-1046$08.50

of the properties of ethanol over wider temperature ranges.

As an approximating function for description of the temperature dependence of density, speed of sound, and isobaric specific heat in ethanol at atmospheric pressure we have chosen a polynomial in powers of $\tau = T/1000$:

$$y = \sum_{i=0}^{n} a_i \tau^i , \tag{1}$$

where y is a function corresponding to the properties ρ^{-1}, W, and C_p; T is temperature in °K as measured by standard MPShT-68. The coefficients a_i of Eq. (1) for ρ^{-1}, W, and C_p were determined by the method of least squares using a Minsk-32 computer.

In processing each set of published data used, a statistical weight was assigned in accordance with the estimated precision of the measurements. The optimum number of coefficients for Eq. (1) was determined by Fischer's criterion.

Density. Data on the density of ethanol at atmospheric pressure and on the saturation curve have been presented in many handbooks [1-3] and are essentially based on values measured by Mendeleev [6] and Yung [8] at temperatures above 273°K. The data available in 1970 were averaged in [3], which offered recommended density values on the saturation curve for temperatures from 253 to 393°K and presented values of Francis equation coefficients for three temperature intervals from 249 to 453°K. The accuracy of the approximation varies with the temperature interval and lies in the range 0.01-0.10%.

In the low-temperature range a wide group of density measurements with uncertainty not exceeding 0.1% were carried out in [9] at temperatures of 159-298°K, while in [10] and [11] relative volume was measured (as compared to volume at 273°K) at 193-293 and 163-283°K, respectively. The most recent (1976) density measurements on the saturation curve at temperatures from 293 to 455°K were carried out by Hales and Ellender [12]. The precision of the ethanol measurements at T = 293-351°K was estimated by the authors to be ±0.02%.

The handbooks [1, 3, 6] present a number of quite precise interpolation equations for the description of specific volume or density, although they cover only a narrow temperature range.

The density values recommended in the so-called alcoholometric tables of [7] for the temperature interval 248-313°K provide reliable information. The accuracy of these values is not stated, but it can be assumed better than ±0.01%.

Preliminary analysis of the available data shows that at $T > 273$°K the most reliable results are those of [7, 12] which furthermore agree well with each other over the region in which they overlap (293-313°K). Deviations do not exceed ±0.1%. At $T < 273$°K we have three basic sets of data [9-11], which agree with each other within ±0.15%, which does not exceed the total uncertainty of the values. While the data of [9] and [11] basically agree within ±0.05%, the data of [10] lie systematically below those of [9] and [11] by 0.12 and 0.09%, respectively. The density values presented in [1, 2] and recommended in [3] for the temperatures of 293.15 and 298.15°K agree with the data of [7, 12] within ±0.01%.

For the final mathematical processing data from [7, 9-12] were used. The data of [10, 11] were corrected using the value of ethanol (100%) density at T = 273°K: $\rho_0 = 0.80623$ g/cm^3 [7] in place of $\rho_0 = 0.8065$ g/cm^3 (99.90%) used in [10, 11]. The density values of [12] at saturation pressure were recalculated to atmospheric pressure (101,325 Pa). The value of the correction did not exceed 0.01%.

The coefficients a_i of Eq. (1) found from this processing are presented in Table 1. Density values calculated from Eq. (1) over the temperature range from fusion (T = 159.05°K [3]) to boiling (T = 351.44°K [3]) at atmospheric pressure are presented in Table 2. Figure 1a shows deviations of the experimental values from the generalized results. For comparison, the deviations of [1-3, 6, 8] are also shown. As is evident from the figure, the present equation describes the original values within the limits of their estimated precision except for the density values of [7] at temperatures of 243-258°K, for which the deviation comprises 0.04-0.06%. This divergence is apparently due to the fact that in composing the tables of [7] at these temperatures less accurate data were used. This is also indicated by the fact that the isobar in a ρ–T diagram constructed with the data of [7] at T = 263°K has a fold, which leads to a discontinuous decrease in density by approximately 0.04% beginning at T = 258°K. A comparison with the most recent data of [13] obtained by extrapolation of experimental

TABLE 1. Coefficients of Eq. (1)

Coefficient a_i	Property		
	ρ^{-1}	W	C_p
a_0	—0,542185381	2663,73851	1,04721599
a_1	36,1455456	—6762,56551	15,6271404
a_2	—343,041903	2921,22328	—107,941020
a_3	1763,03339	18073,7580	310,825090
a_4	—5010,81512	—30701,7452	—241,911840
a_5	7464,40970	—	—
a_6	—4532,41267	—	—

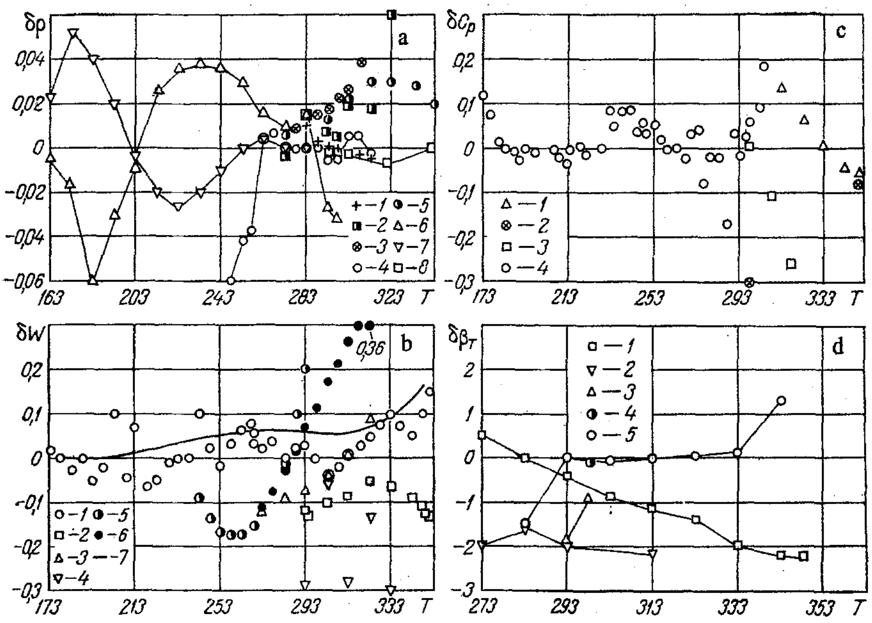

Fig. 1. Comparison of calculated values of density (a), speed of sound (b), isobaric specific heat (c), and isothermal compression coefficient (d) with data of other sources: a) $\delta\rho = (\rho[i] - \rho)\cdot 100/\rho$, %, 1) [1]; 2) [3]; 3) [6]; 4) [7]; 5) [8]; 6) [9]; 7) [11]; 8) [12]; b) $\delta W = (W[i] - W)\cdot 100/W$, %; 1) [15, 16]; 2) [17]; 3) [20]; 4) [18]; 5) [21]; 6) [22]; 7) calculation with equation of [16]; c) $\delta C_p = (C_p[i] - C_p)\cdot 100/C_p$, %, 1) [14]; 2) [3]; 3) [31]; 4) [32]; d) $\delta\beta_T = (\beta_T[i] - \beta_T)\cdot 100/\beta_T$, %, 1) [33]; 2) [34]; 3) [35]; 4) [36]; 5) [37]; i, reference cited.

TABLE 2. Calculated Ethanol Density and Speed of Sound Values

T	ρ	W	T	ρ	W
159,05	0,9073	1715,1	273,15	0,80622	1231,9
163,15	0,9030	1694,9	283,15	0,79778	1196,1
173,15	0,8932	1646,6	293,15	0,78931	1160,9
183,15	0,8840	1599,7	298,15	0,78504	1143,6
193,15	0,8750	1554,0	303,15	0,78075	1126,4
203,15	0,8662	1509,7	313,15	0,77205	1092,3
213,15	0,8575	1466,7	323,15	0,76314	1058,6
223,15	0,8489	1424,8	333,15	0,75397	1025,1
233,15	0,8403	1384,2	343,15	0,74450	991,8
243,15	0,8317	1344,6	351,44	0,73640	964,1
253,15	0,8232	1306,1			
263,15	0,81467	1268,6			

TABLE 3. Experimental Studies of Speed of Sound in Ethanol

Reference	No. of points	Temperature range, deg K	Concentration, % by weight	Uncertainty, %
[15]	18	269—351	99,94	0,04
[16]	24	159,18—268	99,94	0,04—0,06
[17]	11	293—351	99,95	0,04
[18]	6	293—333	—	—
[19]	7	283—343	—	0,5
[20]	6	273—323	99,75	<0,05
[21]	Equation	243—293	100	0,01
[22]	Equation	273—323	100	0,01

TABLE 4. Experimental Studies of Isobaric Specific Heat of Ethanol

Reference	No. of points	Temperature range, deg K	Uncertainty, %
[24]	8	278—314	0,3
[25]	11	196—271	1—1,5
[26]	8	300—330	—
[27]	7	160—298	0,5—1
[28]	20	163—294	1
[29]	9	303—343	—
[14]	5	313—351	0,1—0,2
[30]	5	298—323	0,25—0,28
[31]	41	165—304	<0,20
[32]	3	298—318	<0,30

TABLE 5. Calculated Values of Ethanol Thermodynamic Properties

T	c_p	c_V	h	$\beta_S \cdot 10^5$	$\beta_T \cdot 10^5$
159,05	1,8979	1,417	0	37,47	50,20
163,15	1,9020	1,446	7,79	38,55	50,72
173,15	1,9130	1,499	26,86	41,29	52,71
183,15	1,9259	1,534	46,06	44,21	55,51
193,15	1,9417	1,559	65,39	47,32	58,93
203,15	1,9611	1,582	84,90	50,65	62,80
213,15	1,9848	1,605	104,63	54,21	67,03
223,15	2,0134	1,633	124,61	58,03	71,55
233,15	2,0476	1,667	144,91	62,12	76,32
243,15	2,0880	1,707	165,6	66,50	81,32
253,15	2,1349	1,755	186,7	71,21	86,60
263,15	2,1888	1,811	208,3	76,28	92,20
273,15	2,2501	1,872	230,5	81,73	98,22
283,15	2,3191	1,939	253,3	87,62	104,78
293,15	2,3961	2,011	276,9	94,01	112,00
298,15	2,4376	2,048	289,0	97,41	115,91
303,15	2,4811	2,087	301,3	100,96	120,03
313,15	2,5745	2,166	326,6	108,56	129,01
323,15	2,6762	2,250	352,8	116,94	139,09
333,15	2,7862	2,338	380,1	126,21	150,42
343,15	2,9046	2,432	408,6	136,56	163,12
351,44	3,0089	2,516	433,1	146,11	174,76

data to the saturation curve shows that at T = 195-322°K the latter are elevated by an average of 0.06%. But if we consider that the density measurements in [13] were performed for alcohol 99.84% by weight, then upon recalculation to 100% concentration the agreement improves, since it was shown by special measurements at T = 293.15°K in [14] that a 0.047% by weight water concentration in ethanol leads to a density increase of 0.00015 g/cm^3.

The possible error in the recommended density values calculated with Eq. (1) may be estimated at $\pm$0.01% at T $\geqslant$ 263°K and $\pm$0.05% at T < 263°K.

Speed of Sound. The most complete listing of speed of sound measurements in ethanol is presented and analyzed in [4] and the earlier [15-17]. Since [4] presents some low-accuracy data, those will not be considered further. In [15, 16] a broad range of speed of sound measurements was performed at atmospheric pressure, encompassing almost the entire fusion—boiling temperature range. The authors approximated their measurement results with a third-degree polynomial, to an accuracy within the measurement uncertainty ($\delta W = 0.06\%$).

The goal of the present study was to compose a new interpolation equation with consideration of the most reliable measurements of other authors [17-22], information on which is given in Table 3. It is obvious that increasing the volume of original data used in the processing increases the reliability of the analysis, thus allowing creation of a more accurate equation. The speed of sound values of [17, 20] were recalculated to a 100% alcohol concentration using the technique described in [15].

The coefficients a_i of Eq. (1) for speed of sound are presented in Table 1, while Fig. 1b shows deviations of the experimental values from those calculated with Eq. (1).

As is evident from the figure, the results of the generalization of [16] are elevated relative to the new results over the entire temperature range by an average amount of 0.05%, with the exception of temperatures close to boiling, where the divergence reaches 0.15%.

On the whole, the deviations for the values of [15-22] exceed the uncertainties of each of the authors. This may be due to an inexact knowledge of the content of water or other impurities in the alcohols studied. It has been established [23] that at atmospheric pressure an increase in water content in alcohol by 0.1% by weight increases the speed of sound in the alcohol by 0.66 m/sec on the average. With consideration of this fact and the character of the deviations, it can be assumed that the recommended speed of sound values (Table 2) have an accuracy of ± 0.10-0.15%.

Isobaric Specific Heat. A listing of the basic experimental studies of C_p in pure ethanol is presented in Table 4. Fiock et al. [14] essentially determined specific heat of the liquid phase along the saturation curve C_s. The C_s values were recalculated to atmospheric pressure, the correction not exceeding 0.1%. In [30] C_p values were obtained by extrapolation (from a 94.4% by weight concentration) of specific heat values of aqueous solutions of ethanol. Thus the uncertainty of the data for pure ethanol here may be higher than that presented in Table 4. The C_p values presented in [3] for the fusion and boiling points and $T = 298.15$°K have estimated uncertainties of 0.5, 0.3, and 0.37%, respectively.

The processing utilized data of [14, 24, 25, 27, 28, 30-32], with the C_p values of [14, 30-32] being assigned greater weights.

The coefficients a_i for C_p are presented in Table 1. Figure 1c shows deviations of the experimental C_p values from the results of the generalization. Over practically the entire temperature range, the deviation of the results of [31] is no more than 0.1%, with the exception of several experimental points near the fusion point and a single point at $T = 304$°K, where the deviation is greater than 0.1%, but not more than 0.2%. The deviations of [14, 25, 27, 28, 32] lie within the estimated uncertainty ranges. As for the data of [24, 30] comparison shows that the deviations exceed the measurement uncertainty. While at $T = 298.15$°K the deviation of [30] is -0.02%, with increase in temperature the divergence increases, reaching $+1.2\%$ at $T = 323$°K. The maximum deviations of [24] are $+0.92\%$ at $T = 278$°K and -1.4% at $T = 313$°K. The C_p values of [26, 29] lie systematically above the calculated ones by 1-6%.

Results of a comparison with the most reliable data of [14, 31-32] show that the possible uncertainty of the recommended C_p values (Table 5) can be estimated at $\pm(0.10$-$0.15)\%$.

Calculation of Thermodynamic Properties. In calculating enthalpy values the reference point used was the liquid state at the fusion point under atmospheric pressure ($h_f = 0$). Accordingly, the equation for ethanol enthalpy at atmospheric pressure and temperature T with consideration of Eq. (1) has the form

$$h = h_f + \int_{T_f}^{T} C_p dT = 1000 \sum_{i=0}^{4} a_i [\tau^{i+1} - 0.15905^{i+1}]/(i+1), \tag{2}$$

where a_i are the Eq. (1) coefficients for C_p.

Ethanol enthalpy values calculated with Eq. (2) are presented in Table 5.

Using the interpolation equations obtained for ρ^{-1}, W, and C_p and the thermodynamic relationships of [17] values of C_V, β_S, and β_T were calculated (Table 5). Figure 1d shows a comparison of the calculated β_T values with experimental values from [33-37]. The divergence does not exceed 1-2%, which is within the limits of experimental uncertainty.

NOTATION

T_f, fusion temperature, °K; ρ, density, g/cm^3; W, speed of sound, m/sec; C_V and C_p, isochoric and isobaric specific heats, kJ/(kg·K); β_S and β_T, adiabatic and isothermal expansion coefficients, 1/MPa; h_f and h, enthalpies at fusion point and temperature T, kJ/kg.

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RELATION BETWEEN KINETIC PROPERTIES OF SINGLE CRYSTALS AND OF ORIENTED POLYCRYSTALLINE MATERIALS

E. A. Mityushov, R. A. Adamesku,
and P. V. Gel'd

UDC 539.22

It is demonstrated that the characteristics of a single crystal can be calculated from available data on the properties and the orientation of polycrystalline materials.

Known methods of evaluating the anisotropy of kinetic properties in polycrystalline materials are, as a rule, based on averaging the characteristics of grains over all random orientations. The results obtained in this way do not always agree with experimental data, among others because those methods do not take into account interaction between microstructural components (grains of polycrystalline material). It is therefore definitely important to develop more correct methods for evaluating the anisotropy of kinetic properties in polycrystalline materials.

At the same time, establishment of a proved relation between the kinetic characteristics of polycrystalline materials and those of their single crystals will make it possible to also solve the reverse problem of determining the properties of single crystals from data on

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S. M. Kirov Ural Polytechnic Institute, Sverdlovsk. Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 47, No. 3, pp. 418-421, September, 1984. Original article submitted May 24, 1983.

0022-0841/84/4703-1052$08.50